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NEPTUNIUM SOLUBILITY AND REPOSITORY PERFORMANCE ASSESSMENT, Thomas J.Wolery, Cynthia E. Palmer, and Kevin G. Knauss, Lawrence Livermore National Laboratory, Livermore, CA 94550

Performance assessment (PA) calculations for the potential high-level nuclear waste repository at Yucca Mountain, Nevada have until recently assumed that the transport of dissolved ^{237}Np to the biosphere would be effectively limited by the solubility of NpO_2. This was based on thermodynamic calculations which yield concentrations of dissolved Np on the order of 1×10^{-13} molal. Recent experimental work by Nitsche (1991) and Nitsche et al. (1993) shows that the concentration of dissolved neptunium in J-13 water is controlled at much higher values near 1×10^{-3} molal by equilibrium with $NaNpO_2CO_3{:}3.5H_2O$ (actually of variable composition), Np_2O_5, or both. More recent PA calculations based on these experimental results indicate a problem with the amount of neptunium transported to the biosphere. To begin analyzing the situation, we have carried out some new thermodynamic calculations using the EQ3/6 software. The results show that our understanding of the thermodynamics of neptunium aqueous species and solids is reasonably consistent with the recent experimental results, if one considers that NpO_2 is a "recalcitrant" precipitate at temperatures below at least 90C. That is, the solutions in the experiments appear to be strongly supersaturated with respect to this phase, though its formation is not observed on benchtop time scales. On repository time scales, NpO_2 precipitation may or may not be an effective control on dissolved neptunium. The effect of this mechanism could be evaluated by making a series of experiments in the temperature range 150-250C and from this develop a kinetic model for NpO_2 precipitation that could be extrapolated to lower temperatures and longer times. Another, parallel approach to the PA problem would be to look at the effect of $Np(OH)_4$ as a solubility control in deeper groundwaters with lower redox potentials.

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Contact and Presenting Author:
Thomas J. Wolery
Lawrence Livermore National Laboratory
Earth Sciences Division
L-219, P.O. Box 808
Livermore, CA 94550
(510) 422-5789
Fax: (510) 422-0208
wolery1@llnl.gov

Co-author:
Cynthia E. Palmer
Lawrence Livermore National Laboratory
Isotope Science Division
L-396, P.O. Box 808
Livermore, CA 94550
(510) 422-5693
Fax:
palmer2@llnl.gov

Co-author:
Kevin G. Knauss
Lawrence Livermore National Laboratory
Earth Sciences Division
L-202, P.O. Box 808
Livermore, CA 94550
(510) 422-1372
Fax: (510) 422-1002
knauss1@llnl.gov